5

Air Effluent Monitoring

Arthur H. Biermann
Paula J. Tate

Introduction

Air effluent emissions from facility operations are assessed to evaluate compliance with local, state, and federal regulations and to ensure that human health and the environment are protected from radiological and nonradiological air emissions. LLNL complies with local, state, and federal environmental air quality laws and DOE regulations previously discussed in Chapter 4. In general, LLNL analyzes for most constituents at levels that are far below regulatory standards in order to determine any environmental impact. Air surveillance measurements (see Chapter 4) are also made to help assess LLNL's environmental impact.

Assessment of air effluent emissions is performed by monitoring emissions and/or evaluating potential emissions. Currently, the air effluent sampling program measures only radiological emissions. LLNL has operations with nonradiological discharges; however, permits for these operations are obtained through local agencies having enforcement authority for the Clean Air Act, and monitoring of the effluent is not required. The agencies with oversight for LLNL compliance with air regulations are EPA Region IX, the Bay Area Air Quality Management District (BAAQMD) for the Livermore site and the San Joaquin Valley Unified Air Pollution Control District (SJVUAPCD) for Site 300.

Historically, monitoring of radionuclide air effluents at LLNL has been implemented according to the DOE ALARA (as low as reasonably achievable) policy. This policy is meant to ensure that DOE facilities have capabilities consistent with the types of operations to monitor routine and nonroutine radiological releases, so that the dose to members of the public can be assessed and so that doses are ALARA. In addition, the National Emission Standards for Hazardous Air Pollutants (NESHAPs) 40 CFR 61, Subpart H regulations require that monitoring of facility radiological air effluents must be performed if the potential off-site dose equivalent is greater than 1 μSv/y (0.1 mrem/y), as calculated using the EPA-mandated air dispersion dose model and assuming no emission control devices. For air discharge points that are monitored, the results of the monitoring provide the actual source term for modeling to ensure that the NESHAPs standard, 100 μSv/y (10 mrem/y) total site effective dose equivalent, is not exceeded. Discharges from operations that have the potential to release radionuclides but that are not monitored are also evaluated according to the NESHAPs regulations,

and the corresponding doses are added to those obtained by modeling monitored emissions to determine radiological NESHAPs compliance.

Air effluent monitoring of atmospheric discharge points determines the actual radionuclide releases from individual facilities and processes during routine and nonroutine operations, confirms the operation of facility emission control systems, and can corroborate and aid in the resolution of air surveillance measurement results for the site. (The relationship can also work the other way as well—air surveillance measurements can corroborate effluent monitoring.)

A variety of radioisotopes are used for research purposes at LLNL; these include uranium, transuranics, biomedical tracers, tritium, and mixed fission products. The major radionuclide released to the atmosphere from the Livermore site is tritium. In addition to effluent sampling for tritium, a number of facilities at the Livermore site have air effluent samplers to detect the release of uranium and transuranic aerosols. The air effluent sampling systems described in this chapter apply to stationary and point source discharges. Diffuse, or nonpoint sources, are also monitored to fulfill NESHAPs requirements. Sampling methods to evaluate LLNL diffuse sources are described in Chapter 4, Volume 2. Summary data from these diffuse sources can be found in Chapter 4 of this volume.

Methods

Air effluent monitoring involves the extraction of a measured volume of air from the exhaust of the facility or process and subsequent collection of particles in the extracted volume by filters or of vapors by a collection medium. After collection, the various radionuclides in the sample are measured by appropriate analytical methods.

In all, LLNL maintains 103 radionuclide samplers on air exhausts at 9 facilities at the Livermore site (see **Figure 5-1**). These systems are listed in **Table 5-1** along with the analytes of interest, the type of sampler, and the number of samplers and discharge points monitored. Sampling for particles containing radionuclides is conducted in eight of the facilities; sampling for tritium is conducted in one facility. All sampling systems operate continuously. Samples are collected weekly or biweekly depending on the facility. Most air samples for particulate emissions are extracted downstream of high-efficiency particulate air (HEPA) filters and prior to the discharge point to the atmosphere. Particles in the extracted air are collected on sample filters and analyzed for gross alpha and beta activity. Tritium is collected using molecular sieves. In addition to sample collection for environmental reporting, some facilities have real-time monitors (also listed in **Table 5-1**)

at discharge points to provide faster notification in the event of a release of radioactivity. Analytical results from the continuous samplers are reported as a measured concentration per volume of air, or at the minimum detection concentration (MDC) when no activity is detected. In all cases, the MDC is more than adequate for demonstrating compliance with the pertinent regulatory requirements for radionuclides that are present or may be present in the sampled air. Further details of LLNL air effluent sampling systems are included in Chapter 4 of the *Environmental Monitoring Plan* (Tate et al. 1995).

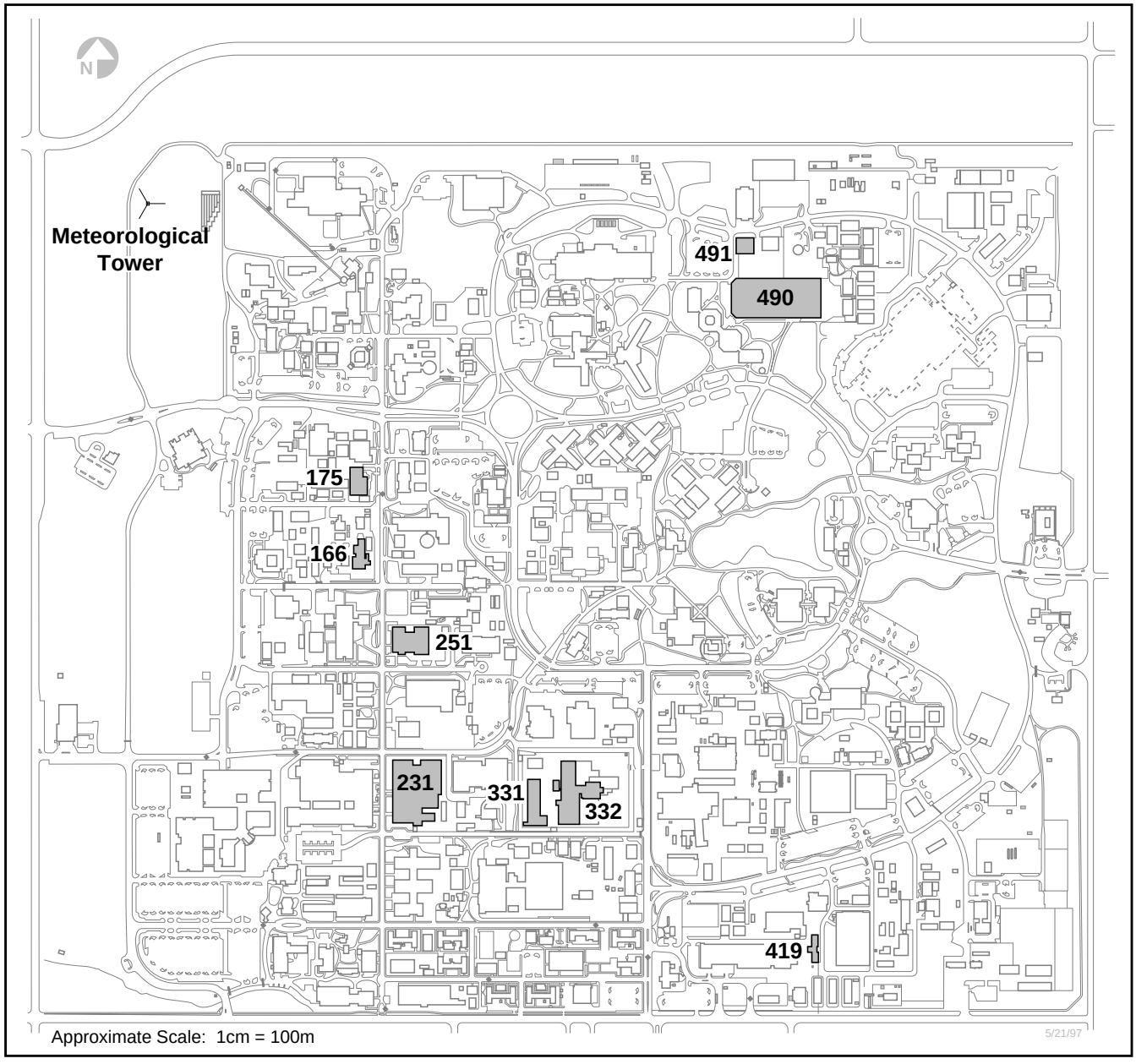

Figure 5-1. Buildings at the Livermore site having air monitoring systems for effluent gas streams.

Table 5-1. Air effluent sampling locations and systems.

Building	Facility	Analytes	Sampler type	Number of samplers	Number of discharge points
166	Pyrochemistry demonstration facility	Gross α, β on particles	Filter	1	1
175	MARS	Gross α, β on particles	Filter	6	6
231	Vault	Gross α, β on particles	Filter	1	1
251	Heavy elements				
	Unhardened area	Gross α, β on particles	Filter	44	55[a]
	Hardened area	Gross α, β on particles	CAM[b]	4	4
		Gross α, β on particles	Filter	4	4
331	Tritium	Tritium	Ionization chamber[b]	4	2
		Gaseous tritium and tritiated water vapor	Molecular sieves	4	2
332	Plutonium	Gross α, β on particles	CAM	12	11
		Gross α, β on particles	Filter	16	11
419	Decontamination	Gross α, β on particles	Filter	2	2
490	Laser isotope separation	Gross α, β on particles	Filter	4	4
491	Laser isotope separation	Gross α, β on particles	Filter	1	1

Note: "CAM" denotes Eberline continuous air monitors.

a Alternate blower system measured by the same sampler.

b Alarmed systems.

The need for continuous air effluent monitoring at other air discharge points that can potentially release radionuclides to the atmosphere is evaluated according to the NESHAPs regulations. The evaluation is based on estimated releases using radionuclide inventories specific to individual discharge points and does not take into account reduction by emission control systems (according to the regulations). The most recent NESHAPs evaluation for LLNL operations is reported in the *LLNL NESHAPs 1996 Annual Report* (Gallegos and Biermann 1997). Many of the existing sampling systems now in place (**Table 5-1**) are not required by regulation; however, LLNL has continued to operate these systems as a best management practice.

Currently, nonradiological emissions (with the exception of beryllium) are permitted through the local air districts, and monitoring of them is not required. The California Air Toxics "Hot Spots" legislation requires facilities to prepare an air toxics emissions inventory and risk assessment, which LLNL has completed. Based on these data, the BAAQMD and the SJVUAPCD have ranked LLNL as a low-risk facility.

Measured Radioactive Air Emissions

This section discusses the radiological air emissions from facilities that have continuously monitored discharge points.

Livermore Site

Tritium emissions from operations in the Tritium Facility (Building 331) account for nearly all the radioactive discharges to the atmosphere from monitored facilities. In 1996, operations in Building 331 released a total of 8.0×10^{12} Bq (210 Ci) of tritium. Of this, approximately 6.7×10^{12} Bq (180 Ci) were released as tritiated water (HTO). The remaining tritium released, 1.2×10^{12} Bq (34 Ci), was elemental tritium gas. The highest single weekly stack emission from the facility was 1.05×10^{12} Bq (28.4 Ci), of which 1.03×10^{12} Bq (27.7 Ci) was tritiated water. The potential dose from tritium gas is approximately 25,000 times lower than the dose from a comparable release of tritiated water. Therefore, the tritiated hydrogen gas did not contribute significantly in calculations of the overall tritium dose. Reduced operations in the facility after 1991 continue to result in lower emissions than in past years (see **Figure 5-2**).

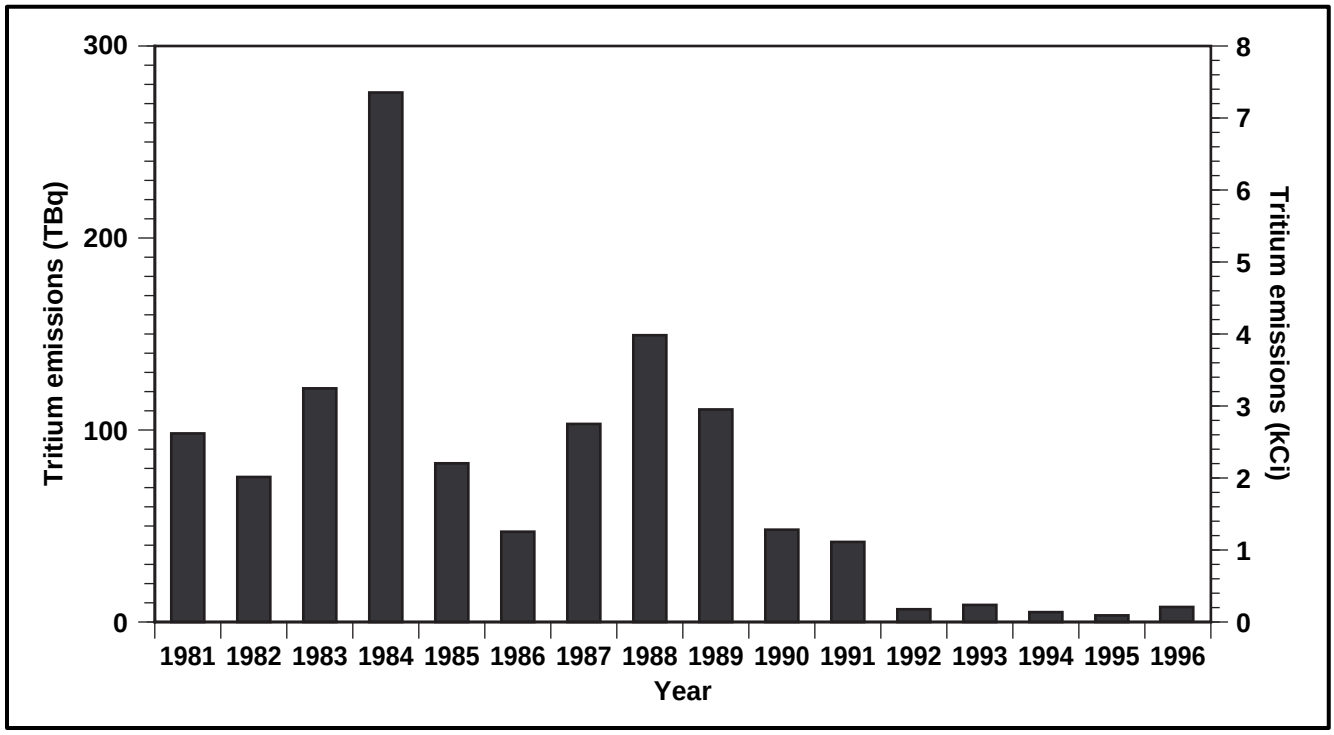

Figure 5-2. Tritium Facility emission (HTO and HT) between 1981 and 1996.

For most of the continuously sampled discharge points having the potential for particulate radionuclide releases, sample results are below the MDC of the analysis. Sometimes as few as 1 to 4 samples (out of 25 to 50 samples per year) have concentrations greater than the MDC. Generally, these few samples having results above the MDC are only marginally above the MDC. Use of zero values for this type of data can be justified based on facility knowledge, the use of HEPA filters in all significant release pathways, and alpha-spectroscopy-based isotopic analyses of selected air-sampling filters. These isotopic analyses have demonstrated the presence of naturally occurring radionuclides, such as radon daughters, e.g., polonium, on air-sampling filters. In addition, because of exhaust configurations at some facilities, the monitoring systems sometimes sample air from the ambient atmosphere in addition to the HEPA-filtered air from facility operations, which gives rise to background atmospheric radioactivity being collected. Because of these considerations, the emissions from such facility operations are reported as zero. Furthermore, even if the MDC values were to be used in calculations of the emission estimates for these facilities, which would be an extremely conservative approach, the total dose to a member of the public attributable to LLNL activities would not be significantly affected.

In 1996, samples from eight emission points at three facilities, three in Building 175, three in 251 (the unhardened area) and two in 419, yielded gross alpha results greater than the MDC on a significant number of the samples collected throughout the year. We use gross alpha as the primary indicator of potential emissions for operations, such as those at Buildings 175, 251, and 419, that involve the use of uranium and transuranic materials. Gross beta results are used as a further corroboration of those gross alpha results having concentrations above the MDC. The gross alpha monitoring concentrations for these buildings ranged from -4.0×10^{-4} Bq / m^3 (-1.1×10^{-14} Ci / m^3) to 3.2×10^{-3} Bq / m^3 (8.5×10^{-14} Ci / m^3). Because of the number of samples with values above the MDC, we have taken a conservative approach and are reporting gross alpha and gross beta measurements as actual emissions. The gross alpha and gross beta emissions for Building 175 were determined to be 3.8×10^3 Bq / y (1.0×10^{-7} Ci / y) and 3.9×10^4 Bq / y (1.1×10^{-6} Ci / y); for Building 251, 1.8×10^4 Bq / y (4.9×10^{-7} Ci / y) and 2.9×10^5 Bq / y (7.9×10^{-6} Ci / y); and for Building 419, 5.9×10^3 Bq / y (1.6×10^{-7} Ci / y) and 9.2×10^4 Bq / y (2.5×10^{-6} Ci / y). **Table 5-2** lists total radiological emissions as determined from the continuous sampling of exhausts for 1996.

We have looked into possible causes of the emissions being reported from Building 419 operations. We found that the physical configuration of the sampling system and faulty seals in the samplers caused some air from the workplace decontamination and decommissioning operations to be sampled by the continuous air samplers. New samplers were installed in October, and since that time no gross alpha or gross beta analyses reported from the new samplers have indicated concentrations above the MDC.

Therefore, the estimated emissions listed in **Table 5-2** are not indicative of emissions from the facility. Actual emissions are now expected to be less than the MDC.

Table 5-2. Measured radiological air effluent emissions for the Livermore site, 1996.

Tritium			
Building	**Facility**	**Elemental, HT (Bq)**	**Tritiated water, HTO (Bq)**
B331	Tritium	1.2×10^{12}	6.7×10^{12}
Gross alpha and gross beta			
Building	**Facility**	**Gross alpha (Bq)**	**Gross beta (Bq)**
B175	MARS	3.8×10^{3}	3.9×10^{4}
B251	Heavy Element	1.8×10^{4}	2.9×10^{5}
B419	Decontamination	5.9×10^{3}	9.2×10^{4}
Total	**Livermore site**	**2.8×10^{4}**	**4.2×10^{5}**

Similarly, the emissions reported for Buildings 175 and 251 have not been confirmed to be emissions from facility operations. As in the case of Building 419, further investigation into the reported emissions is continuing and will include isotopic analyses of selected samples and special air sampling. So it is possible that these emissions from Buildings 175 and 251 are due to naturally occurring, or background, radioactivity, or to the facility exhaust configuration as previously mentioned. In any case, assessment of the gross alpha and gross beta emissions being reported here indicates the radiological dose is only 0.33% of the dose due to all other emissions at the Livermore site and only 0.003% of the NESHAPs standard. Consequently, these reported emissions are not a significant contributor to the dose from Livermore site operations to the public.

Radioactive effluent concentrations from individual discharge points at all facilities being monitored are reported in Chapter 5, Volume 2. Activity concentrations are comparable to the concentrations of gross alpha and gross beta activities as measured by LLNL air surveillance samplers and reported in Chapter 4.

Site 300

Currently, there is no requirement for air effluent monitoring of facilities at Site 300. Air surveillance monitoring is performed for Site 300, and results are reported in Chapter 4.

All Potential Sources of Emissions

This section discusses the evaluation of all sources of radionuclide emissions to air at the Livermore site and Site 300. All discharge points having a potential to release radionuclides to the air are evaluated according to 40 CFR 61, Subpart H of the NESHAPs regulations. This evaluation, performed on an annual basis, uses radionuclide inventories and/or monitoring data along with EPA-accepted release factors for operations and EPA-suggested reduction factors for emission control devices to estimate the potential release for each individual discharge point. Results for 1996 have been published in *LLNL NESHAPs 1996 Annual Report* (Gallegos and Biermann 1997).

Livermore Site Radioactive Emissions

An abbreviated isotope summary of measured and calculated (or potential) emissions for 1996 is presented in **Table 5-3.** Emissions from 41 buildings were evaluated; these buildings, their operations, and effective dose equivalents to a member of the public are listed in Chapter 12 (Radiological Dose Assessment). The estimated release for tritium from both point and diffuse sources was 8.2×10^{12} Bq (220 Ci). Measured emissions from the Tritium Facility account for 97% of the total estimated emissions for tritium. Operations involving tritium at facilities other than the Tritium Facility had estimated releases totaling 1.4×10^{11} Bq (3.7 Ci) during 1996. These releases were assumed conservatively to be HTO. Primary diffuse sources of tritium release are those from Hazardous Waste Management operations at Buildings 514 and 612. These tritium sources have a localized effect. (See Chapter 4.)

A complete isotope listing of calculated emissions appears in Volume 2, Table 5-1.

Site 300 Radioactive Emissions

The estimated radioactive air emissions from Site 300 for 1996 are presented in **Table 5-4**. The estimated uranium releases were point source releases from explosives testing operations at Buildings 801 and 851. The estimated tritium release, 3.5×10^{10} Bq (0.94 Ci), was from diffuse sources, including subsurface tritium contamination. Estimates of a diffuse source of uranium from contaminated soil resuspension were also made. Details of the calculations and assumptions involved in obtaining the soil resuspension estimates are contained in the *LLNL NESHAPs 1996 Annual Report* (Gallegos and Biermann 1997).

Both tritium and uranium contamination are from previous explosives testing.

Table 5-3. Calculated radioactive air emissions from the Livermore site for 1996.

Radionuclide[a]	Calculated emissions[b] (Bq)	Radionuclide	Calculated emissions (Bq)
^{3}H (HTO)[c]	7.0×10^{12}	^{15}O	8.5×10^{10}
^{234}U	9.5×10^{5}	^{238}Pu	6.4×10^{2}
^{228}Th	3.3×10^{5}	^{137}Cs	5.9×10^{6}
^{238}U	2.2×10^{5}	^{32}P	1.9×10^{7}
^{239}Pu	3.9×10^{4}	Gross beta	2.6×10^{5}
Gross alpha[c,d]	3.5×10^{4}	^{90}Sr	1.4×10^{5}
^{13}N	1.6×10^{11}	^{226}Ra	2.2×10^{3}
^{235}U	8.8×10^{3}	^{232}Th	3.9×10^{1}
^{243}Am	1.6×10^{3}	^{3}H (HT)[c]	1.2×10^{12}
^{241}Am	1.1×10^{3}	^{57}Co	3.7×10^{5}

a Radionuclides have been ordered by weighting the emissions according to the inhalation dose rate conversion factor for the isotope.

b Calculated emissions are estimates made according to NESHAPs 40 CFR 61, Subpart H except those noted as measured. Values are considered to be conservative.

c Includes measured emissions from continuously monitored facilities.

d Gross alpha and gross beta activities are reported in inventories where specific isotopic content is not determined.

Table 5-4. Calculated radioactive air emissions from Site 300 for 1996.

Radionuclide[a]	Calculated emissions[b] (Bq)
^{238}U	3.3×10^{9}
^{234}U	3.1×10^{8}
^{235}U	4.3×10^{7}
^{3}H (HTO)	3.5×10^{10}

a Radionuclides have been ordered by weighting the emissions according to the inhalation dose rate conversion factor for the isotope.

b Calculated emissions are estimates made according to NESHAPs 40 CFR 61, Subpart H. Values are considered to be conservative.

Nonradioactive Effluents

The Livermore site currently emits approximately 100 kg/day of criteria air pollutants (nitrogen oxides, sulfur oxides, particulate matter [PM10], carbon monoxide, and lead). The largest sources of criteria pollutants from the Livermore site are surface coating operations, internal combustion engines, solvent operations, and, when grouped together, boilers (oil and natural gas fired).

The estimated releases from exempt and permitted sources of air pollutants at the Livermore site can be compared to the most recent estimated 1995 daily release of air pollutants for the entire Bay Area. For example, the total emissions of oxides of nitrogen released in the Bay Area is approximately 4.8×10^5 kg / day compared to an estimate for LLNL releases of 58 kg / day for the Livermore site (0.012% of total Bay Area emissions). The BAAQMD estimate for reactive organic emissions is 5×10^5 kg / day, versus Livermore site's estimated releases of 30 kg / day (0.006% of total Bay Area emissions) in 1996. **Table 5-5** lists the estimated Livermore site 1996 total airborne releases for criteria pollutants.

Certain operations at Site 300 require permits from San Joaquin Valley Unified Air Pollution Control District. The total estimated air emissions during 1996 from operations (permitted and exempt air sources) at Site 300 are given in **Table 5-5**. Criteria sources at Site 300 include internal combustion engines, boilers, a gasoline dispensing operation, open burning, paint spray booths, drying ovens, and soil vapor extraction.

Table 5-5. Nonradioactive air emissions, Livermore site and Site 300, 1996.

Pollutant	Estimated releases (kg/day)	
	Livermore site	Site 300
Organics/volatile organics	30	1.8
Oxides of nitrogen	58	0.72
Carbon monoxide	10	0.17
Particulates (PM10)	5.5	0.50
Oxides of sulfur	8.1×10^{-1}	4.0×10^{-2}

Environmental Impact

Radioactive air effluents from the Livermore site and Site 300 operations for 1996 are well below levels that should cause concern for the environment or public health according to existing regulatory standards. The doses to the hypothetical maximally exposed members of the public due to measured and potential air emissions, as reported in Chapter 12 (Radiological Dose Assessment), are 0.93 μSv (0.093 mrem) for the Livermore site and 0.33 μSv (0.033 mrem) for Site 300. When compared to the NESHAPs standard of 100 μSv / y (10 mrem / y) and dose from naturally occurring radiation, the estimated doses due to the LLNL radionuclide air emissions reported here are minimal. Nonradioactive air effluents, which are also very small compared to emissions in surrounding areas, are well below standards and do not indicate threats to the environment or public health.